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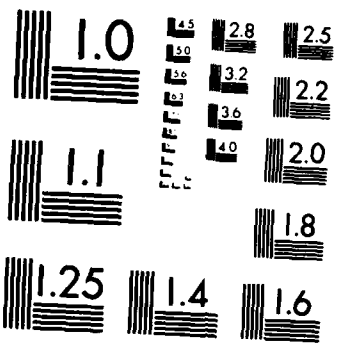

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A pulsed molecular beam valve operated at 100 C was used to produced the supersonically cooled I_2 jet. To achieve the moving grating effect, the pump and probe beams were to be crossed in a plane containing the flow axis, with each beam nearly perpendicular to that axis. The pulsed beam valve produced a broad forward peaked distribution of molecules. The velocity distribution at any given point, however, was mainly along the direction of the line joining that point to the nozzle orifice. This was evidenced by the ~~fluorescence~~ pattern along the laser beam path, which consisted of sharp filaments across the laser beam corresponding to ~~the~~ various hyperfine components of a single rotational transition Doppler tuned into resonance with the laser beam at various angles of inclination. Gain in the planned experiment would be observable only within a narrow range of detuning (obtained by crossing the pump and probe beams), and therefore only a small solid angle of the I_2 jet is useful. ~~A variety of~~ nozzles were tried with the pulsed valve device. Under the best circumstances, an integrated absorption of only about 10% was measured. With the required angular selection, the expected gain would be on the order of ~~10(-2)%~~ .01%.

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(Abstract continued)

Since the stability of the laser was such that even after signal averaging a noise level of the order of 0.1% remained, ~~this~~ precluded our attempt to demonstrate gain ~~in the intended manner~~.

Keywords: Iodine; ——— ; Self pumped; Supersonic flow.

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Self-Pumped Phase Conjugation

in a Supersonically Flowing Medium

AFOSR-84-0226

FINAL REPORT

September, 1987

The intent of this research program was to investigate the feasibility of producing degenerate two wave mixing gain in a noninverted resonant medium with local response. According to theory, two wave mixing gain should be possible if the resulting index grating is shifted in space with respect to the intensity grating produced by the two crossed beams. In the case where the pump and probe beams are degenerate in frequency, the shift between the two gratings results automatically for photorefractive materials which possess nonlocal response. In media with local response, the shift can be brought about by translational motion perpendicular to the planes of the intensity grating. Alternatively, one may observe two wave mixing gain in a stationary medium with local response if the two beams are nondegenerate in frequency.

We chose to study the possibility of generating degenerate two wave mixing gain in a supersonically flowing medium. This approach was selected because the supersonic beam offers a well defined average velocity for the moving medium with an intensity flux not obtainable under thermal conditions. For the resonant medium we chose I_2, because I_2 is a relatively volatile gas with strong transitions in the visible where the required highly stable laser source is available. As a result of cooling in the supersonic expansion process, nearly all I_2 molecules in the jet are in the lowest few rotational levels of the ground vibrational state. This gave us hope that adequate absorption might be realized for the intended demonstration of degenerate two wave mixing gain.

A pulsed molecular beam valve operated at 100°C was used to produce the supersonically cooled I_2 jet. To achieve the moving grating effect, the pump and probe beams were to be crossed in a plane containing the

flow axis, with each beam nearly perpendicular to that axis. The pulsed beam valve produced a broad forward peaked distribution of molecules. The velocity distribution at any given point, however, was mainly along the direction of the line joining that point to the nozzle orifice. This was evidenced by the fluorescence pattern along the laser beam path, which consisted of sharp filaments across the laser beam corresponding to the various hyperfine components of a single rotational transition Doppler tuned into resonance with the laser beam at various angles of inclination. Gain in the planned experiment would be observable only within a narrow range of detuning (obtained by crossing the pump and probe beams), and therefore only a small solid angle of the I_2 jet is useful. A variety of nozzles were tried with the pulsed valve device. Under the best circumstances, an integrated absorption of only about 10% was measured. With the required angular selection, the expected gain would be on the order of 10^{-2}%. Since the stability of the laser was such that even after signal averaging a noise level of the order of 0.1% remained, this precluded our attempt to demonstrate gain in the intended manner.

We did, however, make some very interesting observations relating to two wave mixing of non-Doppler shifted pump and probe beams. We verified for the first time the theory for saturated degenerate two wave mixing absorption in a resonant medium developed by Sargent some years ago. More interestingly, we observed a marked deviation from theory under certain conditions. We have been able to show that the deviation is caused by density inhomogeneities in the I_2 jet.

The details of the two wave mixing absorption measurements are given in the Optics Letters article included in the Appendix. In these

experiments the pump and probe beams were crossed in a plane perpendicular to the flow axis. For the small crossing angle used the relative Doppler shift between the two beams was much smaller than the homogeneous linewidths of the individual hyperfine components. The two beams then interacted with the molecular transitions with essentially the same detunings. For a fixed pump beam intensity, the absorption of the probe beam was measured as its intensity was varied. A representative set of results is shown in Fig. 3 in the attached article. We see that for probe intensities of the order of the saturation intensity or greater the agreement with theory is very good. For weaker probe beam intensities, however, the measured two wave mixing absorption deviates markedly from theory. In the limit of very weak probe intensities, its absorption becomes the same as that for the pump beam alone. We attribute the observed deviation to the wash-out of the density grating in the weak probe limit. That is, when the inherent density inhomogeneities of the medium becomes greater than the amplitude of the density grating created by the pump and probe beams, one should not expect to see any grating effect.

Since the submission of the Optics Letters paper we have conducted additional two wave mixing absorption experiments using a variety of different nozzles. The onset of wash-out of the induced grating was observed to occur over a wide range of probe beam intensities for the different arrangements. In particular, for a highly disturbed jet practically no grating effect was evident for any probe intenisty level. This is illustrated by the data shown in Fig. 1, which were obtained 4 mm from a circular nozzle (0.8 mm diameter) partially blocked by a 0.2 mm thick wire.

We believe the grating wash-out effects observed by us can be used to advantage in determining the optical homogeneity of materials in general, and in studying density variations in gas dynamic systems in particular. We also believe that similar effects should come into play in four wave mixing for sufficiently low probe intensities. That is, for fixed pump intensities in four wave mixing the phase conjugate reflectivity should approach zero for very weak probes. Experiments to verify this prediction are currently underway in our laboratory.

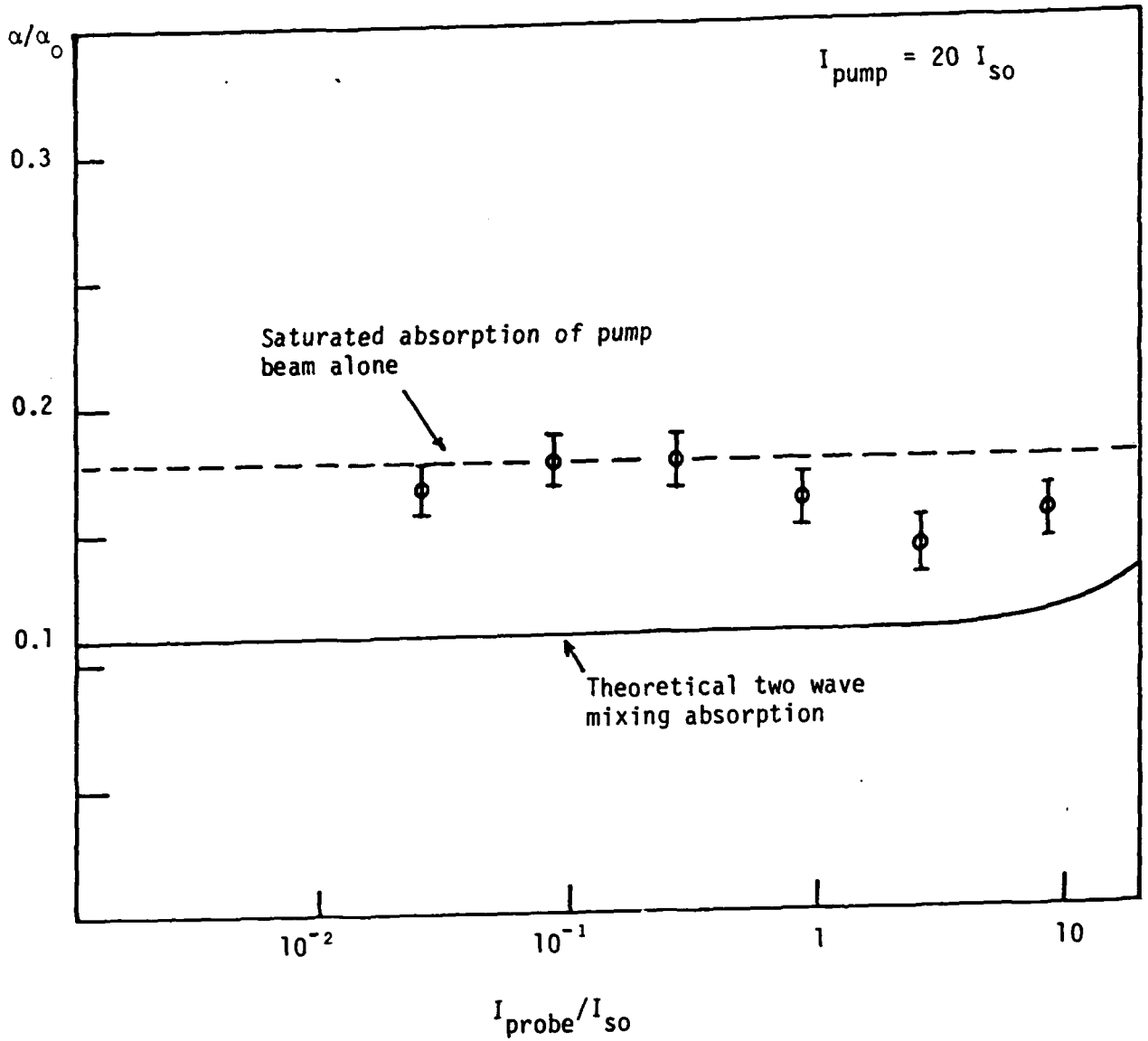
α/α_o
0.3
0.2
0.1
I_pump = 20 I_so
Saturated absorption of pump beam alone
Theoretical two wave mixing absorption
10^-2
10^-1
1
10
I_probe/I_so

FIGURE 1

APPENDIX

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Intensity dependence of two-wave mixing absorption in a resonant medium

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Two-wave mixing absorption in a supersonic beam containing I_2 has been studied. The results are in agreement with theory for probe intensities comparable with the pump intensity. However, at very low probe-beam intensities, the absorption deviates substantially from the predicted value and becomes the same as that for the pump beam alone. We believe that this behavior is caused by density variations in the molecular beam and suggest that this effect can be developed into a tool for probing optical inhomogeneities on a microscopic scale.

Spatial-grating effects resulting from the interference of crossed laser beams have been observed under a variety of conditions.[1] One particular manifestation of these effects is the modification of the attenuation or amplification of the beams themselves. Several mechanisms for this self-diffraction phenomenon were discussed by Vinetskii *et al.*[2] Exchange of power between the two beams because of one of these mechanisms (which was thermal in origin) has been observed.[3] Starting from a different formalism, Sargent[4] has analyzed the effect of spatial gratings on saturation in a resonant medium. We have obtained confirmation of Sargent's results by using a supersonic jet of I_2 as the resonant medium. The agreement, however, did not hold for small probe-to-pump intensity ratios. A possible explanation for the discrepancy is proposed.

A pulsed-beam device maintained at 100°C was used to produce the supersonic I_2 beam. I_2 vapor was generated in an external reservoir kept at 90°C and was carried into the beam-valve chamber by He at a pressure of 800 Torr. The gas mixture was expanded through a circular nozzle with a diameter of 0.5 mm and a length of 1 mm. The duration of the pulses was approximately 1 msec, and a typical pulse repetition rate of 1 sec^{-1} was used. When a single-frequency dye-laser output was used to excite a single rotational transition in the $B(^3\Pi^+_{ou}) \leftarrow X(^1\Sigma^+_g)$ system, fluorescence in the form of a line as observed in a direction normal to both the laser-beam and the molecular-beam axes moved along the laser beam as the frequency of the dye laser was tuned. On close examination, the fluorescing line, which subtended an angle of approximately 60° at the nozzle opening, consisted of numerous unevenly spaced filaments running across the laser beam in a fanlike arrangement. The extensions of these filaments all appeared to converge at the nozzle orifice. Since it is well known that each transition in I_2 is composed of a large number of hyperfine components,[5,6] these filaments can be explained if the local gas velocity at each point along the laser beam has a reasonably well-defined direction, namely, along the line joining the point to the nozzle opening. Thus each filament represents a given hyperfine component or a group of nearby hyperfine components excited by the laser. When the laser beam was made sufficiently thin ($\ll$1 mm), the filaments extended beyond the width of the laser beam in the downstream direction. The length of the filaments was in accord with that calculated from the velocity of the gas flow and the radiative lifetime of the vibrational level of the $B(^3\Pi^+_{ou})$ state excited.[7]

With the frequency-stabilized dye laser tuned to the $P(7)$ transition in the $(22 \leftarrow 0)$ band,[8] saturation measurements were made with a 0.25-mm-diameter beam 5 mm away from the nozzle opening. For the $P(7)$ transition there are 21 hyperfine components. Consider first the interaction of one of these components with the laser field. Under our experimental conditions, the homogeneous linewidth of the component is of the order of 1 MHz. Therefore, for a given hyperfine component, only molecules with axial (with reference to the laser beam) velocities within a small range will resonantly interact with the field. Since the total angular spread of the molecular-beam cloud is quite large, it is reasonable to assume that the same density of molecules participates in the interaction, regardless of the amount of detuning on a given hyperfine component. Thus one should be able to write the small-signal path-integrated absorption that is due to the ith hyperfine component as

$$A_{0,i} = \frac{c\sigma_i N_i \gamma_i R_i}{v\omega_i} \int \frac{d\Delta}{1+\Delta^2} = \frac{\pi c \sigma_i N_i \gamma_i R_i}{v\omega_i}. \qquad (1)$$

Here N_i is the density, σ_i the absorption cross section, ω_i the line-center frequency, and γ_i the half-width in angular units, all referring to the ith hyperfine component. In addition, v is the average magnitude of molecular velocity, and R_i is the distance of the filament identified with the ith hyperfine component from the nozzle opening. The integration is over all possible values of Δ, which is the frequency detuning in angular units normalized to the half-width of the transition.

Similarly, the saturated path-integrated absorption

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that is due to the ith hyperfine component can be written as

$$A_i = \frac{c\sigma_i N_i \gamma_i R_i}{v\omega_i} \int \frac{d\Delta}{1 + \Delta^2 + (I/I_{s,i})} = \frac{A_{0,i}}{\sqrt{1 + (I/I_{s,i})}}, \quad (2)$$

where I is the intensity of the laser beam and $I_{s,i}$ the saturation intensity at line center for the ith hyperfine component. For the $P(7)$ transition, all the hyperfine components have absorption cross sections within about 15% of the mean.[9] Therefore it should be a good approximation to use an average line-center saturation intensity I_{s0} and write for the total saturated absorption

$$A = \frac{A_0}{\sqrt{1 + (I/I_{s0})}}, \quad (3)$$

where $A = \sum A_i$ and $A_0 = \sum A_{0,i}$. The measured saturated absorptions are plotted in Fig. 1 for $I_{s0} = 4.4$ W cm^{-2} along with the calculated curve using Eq. (3). Each point is the average over 30 pulses, and the error bars give a measure of the noise level on the averaged signals. It is seen that a good fit is obtained for this particular choice of I_{s0}, justifying the approximation made in Eq. (3).

Saturated absorption in two-wave mixing was investigated by using the arrangement shown in Fig. 2. The pump and probe beams crossed in a plane parallel to the nozzle plate and 5 mm from it, with their polarization vectors perpendicular to the plane. The crossing region was positioned directly in front of the nozzle. In this way any Doppler shift between the two beams was negligible. That is, any given molecule interacted with the two beams with essentially the same detuning. The pump and probe beams had diameters of 0.25 and 0.17 mm, respectively, and crossed at an angle of 20 mrad. Under these conditions, good overlap between the pump and probe beams was obtained for the entire interaction region between the probe beam and the molecular-beam cloud. Outside the region of overlap, any absorption of the probe beam was negligible.

The theory of saturated absorption for two-wave

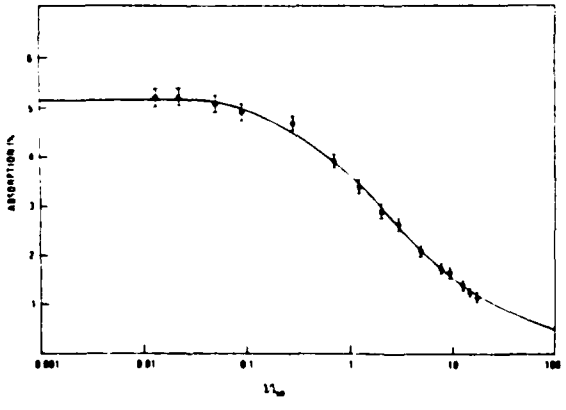

Fig. 1. Measured single-beam saturated-absorption data plotted against the curve given by Eq. (3) when $I_{s0} = 4.4$ W cm^{-2} is used.

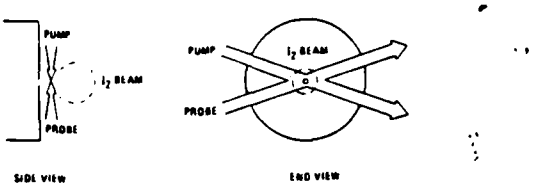

Fig. 2. Experimental arrangement for saturated two-wave mixing absorption studies. The side view is along the bisector of the two laser beams, and the end view is facing into the molecular-beam axis. The tapered arrows in the side view are meant to show that the two beams are going into the paper.

mixing in a resonant medium has been developed by Sargent.[4] It was shown that, in the absence of any relative Doppler shift between the probe and pump beams, the saturated absorption coefficient for the probe as a function of detuning Δ (same for both probe and pump) is given by

$$\alpha(\Delta) = \sigma N S(\Delta), \quad (4)$$

with

$$S(\Delta) = \frac{I_{s0}}{2I_1}\left[1 - \frac{1 + (I_2' - I_1')}{[1 + 2(I_1' + I_2') + (I_1' - I_2')^2]^{1/2}}\right] \quad (5)$$

and

$$I_j' = \frac{1}{1 + \Delta^2}\frac{I_j}{I_{s0}}, \quad (6)$$

where the subscripts 1 and 2 refer to probe and pump, respectively. If we again assume that the same saturation intensity applies to all the hyperfine components, the net two-wave mixing absorption experienced by the probe is given by

$$A_{TWM} = A_0 \int S(\Delta) d\Delta. \quad (7)$$

We note that, although Sargent's results were obtained for counterpropagating beams, they are equally valid for beams crossing at arbitrary angles, so long as a sufficiently large number of grating periods are involved.

The measured probe-beam absorptions at a fixed pump-beam intensity of $I_2 = 25I_{s0}$ for probe-beam intensity variations over nearly 4 orders of magnitude are shown in Fig. 3. The laser frequency was tuned so that molecules in the central portion of the jet were excited. The solid curve is that obtained when Eq. (7) is numerically evaluated. The value for the saturation intensity I_{s0} used in both plotting the experimental points and evaluating the integral is the one determined from the data shown in Fig. 1. No adjustable parameter has been used anywhere in the presentation of the figure. We note that the agreement between the experimental results and theory is quite good for probe intensities of the order of the saturation intensity or greater. From Eq. (5) we see that, for a fixed detuning and for $I_2' \gg 1$, probe-beam absorption should increase from $1/(I_2')^2$ times the small-signal absorption for $I_1' \ll 1$ to $1/(2I_2')$ times the small-signal

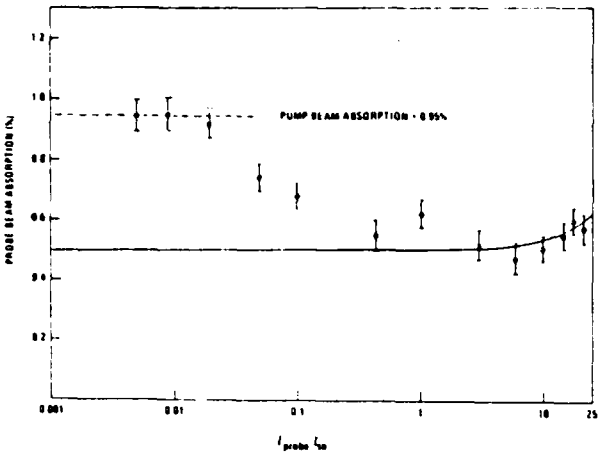

Fig. 3. Measured probe-beam absorption versus its intensity for a small-signal absorption of 5.5% and $I_{pump} = 25I_{s0}$. The solid curve is given by Eq. (7), and the dashed line indicates the single-beam saturated absorption of the pump.

absorption for $I_1' = I_2'$; that is, the population-density grating effect is the most pronounced for very small probe-to-pump intensity ratios. As can be seen from the calculated curve in Fig. 3, even after averaging over detuning has been performed, this behavior is still expected. This predicted increase in absorption when the probe-beam intensity approaches the pump-beam intensity appears to be supported by our experimental results.

For probe intensities much smaller than the saturation intensity, the experimental data deviated substantially from the predicted behavior. In fact, for very low probe-beam intensities the measured absorption became the same as that for the pump beam alone. In other words, the grating effect appeared to be absent for very small probe-to-pump intensity ratios. We believe that this behavior is caused by the presence of density inhomogeneities in the molecular beam. In fact, if there are density variations comparable with or smaller than the grating period in scale and comparable with or larger than the amplitude of the induced grating in magnitude, one would expect a washout of the grating effect. In our experiment the data would imply the presence of spatial-density variations of the order of 1% of the average density.

The washout of the grating effect in two-wave mixing because of inhomogeneities in the entity responsible for the formation of the induced grating can potentially be used as a microprobe for the uniformity of that entity. Knowledge about the onset of washout would provide a measure of the magnitude of the nonuniformity. Information on the scale of the inhomogeneity may be obtained by varying the crossing angle of the probe and pump beams and hence the grating period. By rotating both beams, one can, in principle, detect any anisotropy of the spatial variation.

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